al. For a sample with ten 2000 Å layers of NbN separated by nitrogen-rich NbN, critical current densities about twice that of a corresponding unlayered sample have been observed.

VI. SUMMARY AND CONCLUSIONS

At Gulf General Atomic, thin films of NbN have been grown by high-vacuum vapor codeposition of niobium and nitrogen at rates of up to several hundred Å/sec. Such films have transition temperatures of 12.5–13 °K and critical current densities in excess of 10^6 A/cm^2. The superconducting properties of these films have been investigated for a variety of fabrication techniques, and it has been concluded that the depression of T_c from bulk value is probably due to the inclusion of impurities in the film from the nitrogen source. Although both diatomic nitrogen and ammonia were used as nitrogen sources, only the ammonia proved effective in forming NbN.

It has been shown that NbN with moderate transition temperatures can be fabricated in thin-film form at high rates (> 100 Å). If the material can be shown to have critical fields comparable to those of sputtered NbN films, the techniques described here should allow for the production of useful high-field superconducting magnet material. The substrate temperatures during fabrication are low enough to allow high-purity copper to be used as the substrate, thus providing thermal stabilization.

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Microwave Loss in Ultralow-Linewidth Substituted Garnets

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Linewidth measurements from 77 to 300 °K and room-temperature off-resonance effective linewidth data at 9 GHz show that the microwave losses in ultralow-loss Ca-V-In substituted YIG and Ca-Ge-In-YIG are completely resolvable in terms of known processes and that only a small part of the loss is due to anisotropy broadening. For the lowest linewidth of 2 Oe observed to date (Ca-Ge-In-YIG), about 0.5 Oe is due to rare-earth impurities, 0.9 Oe is from porosity broadening, and 0.2 Oe is from anisotropy. The remaining 0.4 Oe contribution is comparable to expected "intrinsic" single-crystal linewidths.

As a result of Ca-V-In substitutions in YIG, the crystalline anisotropy field can be reduced to very low values with a corresponding reduction in the FMR linewidth in polycrystals.[1] These and related substitutions have resulted in linewidths as low as 2.0 Oe.[2,3] The low linewidths have generally been attributed to inhomogeneous broadening due to the random orientation of crystallites, so-called anisotropy broadening. In fact, the linewidth data were often used to actually estimate the crystalline anisotropy field H_A for the material. Data taken on polycrystalline YIG,[4,5] on the other hand, have indicated that at least 1 Oe of the linewidth is due to some as-yet undertermined source. The present data on ΔH vs temperature at 9 GHz and 77–300 °K and off-resonance effective linewidth[6] at room temperature show that neither of these possibilities applies to the ultralow-linewidth garnets considered here. A careful analysis of the linewidth temperature dependence of ΔH shows that only a very small linewidth contribution is attributable to anisotropy. Moreover, the entire linewidth can be satisfactorily accounted for by using known and well-understood relaxation mechanisms. The results indicate an ultimate polycrystalline linewidth limitation of about 0.5 Oe. The presence of residual porosity ($\approx$0.05%) pushes the practical linewidth limit up to about 1.2 Oe.

Three different materials were examined, two Ca-V-In substituted YIG samples with linewidths of 4.4 and 2.2 Oe, and a Ca-Ge-In substituted garnet[7] with a linewidth of 2.0 Oe. Basic magnetic data on these materials are given in Table I. Linewidths vs temperature for the three samples are given in Fig. 1. For all three, ΔH increases substantially in going from room temperature down to 77 °K, but only for the YCIVF45 is the increase in agreement with that predicted for anisotropy broadening,[8]

$$\Delta H_{\text{anis}} = (8\pi\sqrt{3}/21)(H_A^2/4\pi M), \tag{1}$$

where H_A is the crystalline anisotropy field and $4\pi M$ is the saturation induction. From the data in Ref. 9, $\Delta H_{\text{anis}}^{(77\,^\circ\text{K})}/\Delta H_{\text{anis}}^{(300\,^\circ\text{K})}$ is estimated to be 9.6, 20.0, and 68.0 for YCIVF45, YCIVF40, and YCIGeF, respectively. The observed increase for YCIVF45 is a factor

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TABLE I. Basic magnetic data for the three substituted garnets, YCIVF45, YCIVF40, and YCIGeF.

Sample	Composition[a] x	y	$4\pi M$ (300 °K) (G)	T_c (°C)	ΔH_{exp} (Oe)
YCIVF45	0.45	0.40	1250	200	4.4
YCIVF40	0.40	0.60	1060	135	2.2
YCIGeF[b]			1170	115	2.0

[a] $(Y_{3-2x}Ca_{2x})(Fe_{5-x-y}V_xIn_y)O_{12}$ for YCIVF, YCIGeF has composition $Y_{2.6}Ca_{0.4}Fe_{3.3}In_{0.7}Ge_{0.4}O_{12}$.

[b] Obtained from G. Winkler, Philips Laboratories, Hamburg.

of 8.2, in reasonable accord with Eq. (1). The observed increase for the ultralow-loss materials, however, is only a factor of 6.6 and 5.7 for YCIVF40 and YCIGeF, respectively. These increases are much lower than expected for anisotropy broadening. It is clear, therefore, that anisotropy is not a major contributor to the linewidth for these low-loss materials.

To determine what processes actually *do* contribute to the linewidth in YCIVF40 and YCIGeF, additional effective linewidth data have been obtained and a detailed analysis of the temperature data in Fig. 1 has been carried out. The linewidth increase with temperature T was described analytically by an impurity contribution which varied as T^{-2} and the anisotropy contribution in Eq. (1) where H_A and $4\pi M$ increase with decreasing temperature as described in Ref. 9. Seiden's data on rare-earth impurity relaxation[10] have shown a T^{-2} dependence in the range $77 < T < 300$ °K. The actual linewidth data $\Delta H(T)$ were fitted to a working equation,

$$\Delta H_{obs}(T) = \Delta H_{imp}\left(\frac{300}{T}\right)^2 + \Delta H_{anis}\left(\frac{(H_A^2/4\pi M)_T}{(H_A^2/4\pi M)\,300\ °K}\right) + \Delta H_{res}, \quad (2)$$

where ΔH_{imp}, ΔH_{anis}, and ΔH_{res} refer, respectively, to the impurity, anisotropy and residual linewidth contributions at 300 °K. For purposes of analysis, contributions to the loss other than impurity and anisotropy broadening have been lumped into a temperature-independent ΔH_{res}. Equation (2) was applied to the data at 300 °K, 77 °K, and some intermediate temperature to generate a set of three simultaneous equations, which were solved to get ΔH_{imp}, ΔH_{anis}, and ΔH_{res}. A simple check on the validity of Eq. (2) in describing the linewidth temperature dependence was made by varying the intermediate temperature used in this analysis and examining the scatter in the different ΔH_{imp}, ΔH_{anis}, and ΔH_{res} values so obtained. The scatter was generally on the order of 0.1 Oe, which indicates that Eq. (2) is a valid description of the physical situation.

Strictly speaking, Eq. (2) should contain a temperature function multiplying ΔH_{res} which reflects the proper temperature dependence for the particular processes to which ΔH_{res} is attributed. However, the temperature dependences for likely contributions to ΔH_{res}, such as porosity broadening or thermally activated intrinsic processes, are much weaker than those for impurity relaxation or anisotropy broadening and have little effect on the results. For example, the inclusion of a factor $4\pi M(T)/4\pi M(300\ °K)$ multiplying ΔH_{res} in Eq. (2) modified the calculated linewidth contributions by less than 0.1 Oe in all cases.

The numerical results which came out of the analysis are tabulated in the first three columns of Table II. The rare-earth impurity contribution to the linewidth decreases from 0.88 Oe for YCIVF45 down to 0.35 Oe for YCIGeF. Based on a purity of 99.99% for the Y_2O_3 starting material used to produce the YCIVF samples and a breakdown of the constituent rare-earth impurities by West,[11] an impurity linewidth on the order of 0.5 Oe is expected for the YCIVF40. The 0.58 Oe of impurity linewidth which comes out of the analysis is very close (fortuitously) to this value. The YCIGeF material shows an even smaller impurity linewidth. This 0.35 Oe value is a factor of 3 lower than the ΔH_{imp} estimated from the Y_2O_3 impurity content given in Ref. 2, based on Seiden's impurity linewidth measurements.[10] Consequently, it appears that the Philip's starting materials are somewhat better than expected.

The anisotropy linewidth for YCIVF45 is almost exactly the same as expected from anisotropy measurements on single crystals of the same nominal composition.[9] The single-crystal data indicate an anisotropy field H_A of 32 Oe. The present analysis gives $H_A = 33.3 \pm 0.8$ Oe. This close agreement implies that the present approach is useful in giving accurate estimates of H_A. In the case of YCIVF40, reliable single-crystal data are not available. The indium content for YCIVF40 is twice that for the closest single-crystal composition in Ref. 9. A crude extrapolation from Ref. 9 does give a value $H_A \approx 23$ Oe, which is much too large to fit the data. Although the errors are relatively large for the YCIGeF material, the anisotropy contribution to the linewidth is clearly very small, less than 0.1 Oe.

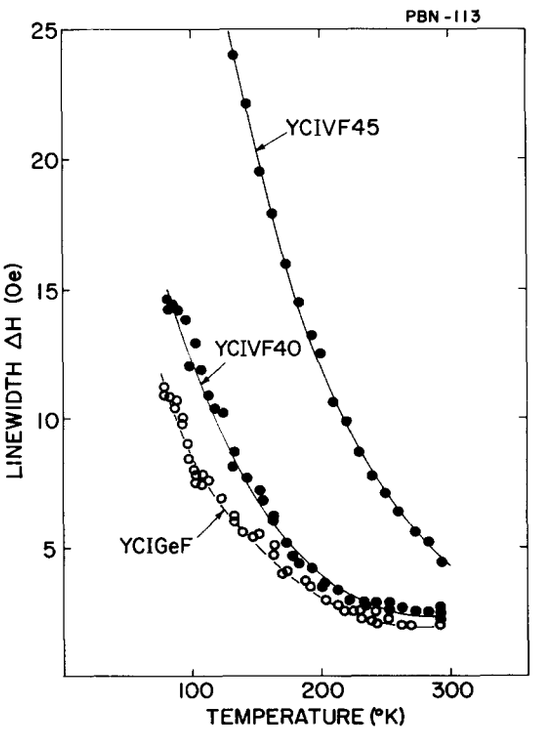

FIG. 1. Resonance linewidth ΔH at 9 GHz as a function of temperature for three substituted garnets, YCIVF45, YCIVF40, and YCIGeF.

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TABLE II. Room-temperature linewidth contributions for three substituted garnets, YCIVF45, YCIVF40, and YCIGeF.

Sample	ΔH_{imp} (Oe)	ΔH_{anis} (Oe)	ΔH_{res} (Oe)	ΔH_{eff} (Oe)	ΔH_{por} (Oe)	ΔH_{int} (Oe)
YCIVF45	0.88± 0.06	2.37± 0.11	1.15± 0.06	1.3± 0.3	0.73± 0.42	0.42± 0.36
YCIVF40	0.58± 0.16	0.22± 0.11	1.40± 0.06	1.0± 0.3	0.98± 0.52	0.42± 0.46
YCIGeF	0.35± 0.16	0.05± 0.04	1.60± 0.12	1.1± 0.3	0.85± 0.58	0.75± 0.46

The room-temperature residual linewidth which comes out of the analysis ranges from about 1.2 to 1.6 Oe. Let us write ΔH_{res} as a combination of two terms, ΔH_{por} and ΔH_{int}, where subscripts por and int refer to porosity and intrinsic. "Intrinsic" includes all processes (as yet unidentified) not explicitly included by the other terms[12]:

$$\Delta H_{res} = \Delta H_{por} + \Delta H_{int}. \tag{3}$$

These contributions may be evaluated explicitly, by taking off-resonance effective linewidth results into account. As discussed previously, neither anisotropy nor porosity scattering relaxation contribute to the off-resonance effective linewidth ΔH_{eff},

$$\Delta H_{eff} = \Delta H_{imp} + \Delta H_{int}. \tag{4}$$

Experimental values of ΔH_{eff} for the three materials, obtained using the technique in Ref. 13, and values of ΔH_{por} and ΔH_{int} calculated from Eqs. (3) and (4) are listed in Table II.

The porosity linewidth is related to the effective porosity P_{eff} through

$$\Delta H_{por} = 1.5 \times 4\pi M P_{eff}. \tag{5}$$

From Eq. (5) and the results in Table II, the effective porosities for YCIVF45, YCIVF40, and YCIGeF are 0.04, 0.06, and 0.05%, respectively. Such low porosities are typical of values attainable by careful material preparation using standard sintering techniques. Of course, it should be noted that these effective porosities include second-phase content as well as air pores. These results show that residual porosity broadening represents a major part of the linewidth in these ultralow-loss materials. It is unlikely, moreover, that significantly higher densities can be attained with known ceramic procedures. Consequently, it appears that no major reductions in this linewidth contribution can be accomplished, except possibly by reducing the saturation magnetization.

The remaining linewidth is tabulated in the last column of Table II, under the label ΔH_{int} for lack of any better designation. This is the remaining loss after contributions due to anisotropy broadening, rare-earth impurity relaxation, and porosity have been subtracted. The linewidth values, 0.45, 0.45, and 0.75 Oe for YCIVF45, YCIVF40, and YCIGeF, respectively, are still somewhat larger than the room-temperature 0.22-Oe linewidth reported for ultrapure single-crystal YIG.[14] This intrinsic YIG linewidth, however, was reported to vary inversely with $4\pi M$. If the reduced $4\pi M$ for the present low-loss materials ($4\pi M \approx 1750$ G for YIG) is taken into account, the expected intrinsic linewidths are about 0.35 Oe. This value is quite close to the ΔH_{int} values in Table II for YCIVF45 and YCIVF40 and within the experimental error for the YCIGeF.

In summary, it appears that the linewidth in state-of-the-art ultralow-loss substituted YIG is completely resolvable in terms of well-established relaxation processes. A detailed analysis of the linewidth variation with temperature, combined with effective linewidth data, indicates that the 2-Oe linewidth breaks down into 0.5 Oe of impurity relaxation, a few tenths Oe of anisotropy broadening, about 0.9 Oe due to a residual 0.05% porosity, and approximately 0.4 Oe due to intrinsic processes of the sort used to explain single-crystal YIG relaxation. The impurity contribution could be removed, in concept, simply by using more refined Y_2O_3 starting material. The anistropy contribution could also be eliminated by proper substitution levels. The residual porosity of 0.05% is probably a factor which must be lived with, barring the advent of radically improved ceramic processing techniques. As a result, the lowest attainable polycrystalline linewidth should be on the order of 1.2 Oe, depending on the magnetization, a further reduction of 40% from the present state-of-the-art 2.0-Oe linewidths.

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